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Local field in LiD polarized target material

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Abstract

We have experimentally studied the first and the second moments of D, ^{6}Li and ^{7}Li ($I > \frac{1}{2}$) NMR lines in a granulated LiD-target material as a function of nuclear polarizations and the data has been compared with a theory elaborated by Abragam, Roinel and Bouffard for monocrystalline samples. The experiments were carried out in the large COMPASS twin-target at CERN. The static local magnetic field of the polarized nuclei was measured by frequency shift between the NMR-signals in the two oppositely polarized cells and lead to the first moment, whereas the investigation of the second moment was done through Gaussian approximation. The average field magnitude in granulated material was estimated 20% larger than the value given by the calculations for monocrystalline samples of cylindrical shape. The second moment shows a qualitative agreement with the theory but it is slightly larger at the negative than at the positive polarization. In a polarized mode, the moments depend on the saturated microwave field.

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1. Introduction

The dynamic nuclei polarization (DNP) is a resonance method to obtain a high nuclear polarization. DNP efficiency depends on the chemical consistence of a material, on the performance of the ^{3}He/^{4}He dilution refrigerator, as well as on the uniformity of the magnetic and microwave (MW) fields. The record deuteron average polarization of 57% has been reached in the granulated LiD-material of the COMPASS target at CERN, but the measurements showed a $\pm 5\%$ longitudinal deviation of polarization, besides the maximum of the negative polarization was found to be 5% less than the positive one. For further improvement of the target performance, we

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analysed the reasons for nonuniformities through the first and second moments of D, ^{6}Li and ^{7}Li NMR lines. Whenever possible the data were compared with the theory [1,2] for the monocrystalline samples.

2. The local field in LiD

COMPASS polarized twin-target at CERN consists of the two independently and oppositely polarized cells, each cell 60 cm long and 3 cm in diameter, filled with irradiated ^{6}LiD-granulated material. The nuclear polarization was measured by ten commercial "Liverpool" Q-meters [3] connected to probing NMR coils equally distributed along the upstream and downstream cells. The coils were made in the saddle shape of 8 cm length. The receiver circuits of the Q-meters were permanently tuned to the $\nu_0 = 16.38$ MHz and fed by a RF synthesizer, the frequency of which was scanned at 1000 frequency steps within the 50 kHz bandwidth across the frequency center.

All of the nuclear species in the LiD target material can be polarized. The local field of the polarized nuclear moments shifts the Larmor frequencies of NMR spectra. Fig. 1 shows the typical shift between D and ^{7}Li signals in the oppositely polarized cells taken in the resonant fields $H_k = \nu_0/\gamma_k$, where γ_k is the gyromagnetic ratio of the species (see Table 1). The local field represents the linear combination of the nuclear polarizations, the spin densities and the spins. For a long cylindrical monocrystal, we have [1]:

$$m_{1k}^{\mathrm{up,d}} = \left(\frac{2\pi}{3}\right) h\Big(3\gamma_k N_k^{\mathrm{up,d}} I_k P_k^{\mathrm{up,d}} + 2\sum_{i \neq k} \gamma_i N_i^{\mathrm{up,d}} I_i P_i^{\mathrm{up,d}}\Big) \quad (1)$$

where m_{1k}^{up} and m_{1k}^{d} are the theoretical first moments (in Gauss) of the upstream and downstream targets, index k stands for D, ^{6}Li, ^{7}Li and H spins, respectively. I_k is the spin of the k-nucleus, N_k is the k-spin density and h is the Planck's constant. $P_k^{\mathrm{up,d}}$ is the polarization of the corresponding nucleus calculated over the deuteron polarization using the Equal Spin Temperature (EST) concept. The EST validity in LiD was established in Ref. [4]. We measured the difference

$$m_{1k}^{\mathrm{exp}} = \alpha_{1k}^{\mathrm{exp}}(m_{1k}^{\mathrm{up}} - m_{1k}^{\mathrm{d}}) + C_k \quad (2)$$

where m_{1k}^{exp} (in Gauss) denotes the shift between the maxima of the two NMR spectra in oppositely polarized cells, C_k is the contribution of the nonuniform field and the local field of the electron spins in the frozen mode (C_k is adjusted to zero at zero polarization), and $\alpha_{1k}^{\mathrm{exp}}$ is the normalization between experiment and theory. The spin density was determined by the chemical composition and the weighing of the material. The mass-spectrometric analysis of the material revealed that it contains an atomic fraction of 0.953 for ^{6}LiD, 0.0440 for ^{7}LiD and 0.003 for ^{6}LiH. The upstream spin density is given in Table 1; the downstream spin density is 1.087 times greater than the upstream one, due to a denser packing of the material.

The maximum shift of the local field was of the order of the earth magnetic field. Fig. 2 shows the field shift of D and ^{6}Li averaged over

Table 1

k	I_k	γ_k (Hz/G)	Field H_k (T)	N_k^{up} 10^{22} (sp/cm^3)
D	1	653.6	2.506	2.99
^{6}Li	1	626.6	2.614	2.86
^{7}Li	3/2	1654.8	0.9898	0.132
H	1/2	4257.7	0.3847	0.00898

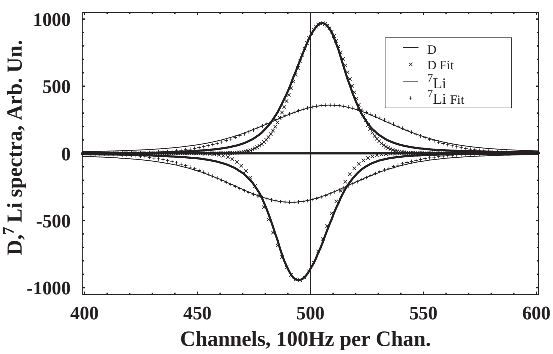

Fig. 1. D, ^{6}Li, ^{7}Li spectra and Gauss fit of D and ^{7}Li; $P_D = 39.2\%$ (top), and $P_D = -36.9\%$ (bottom).

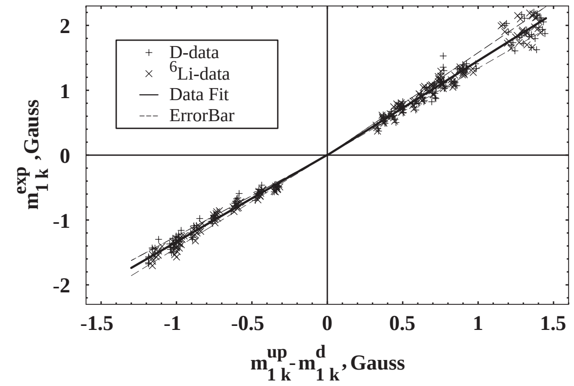

Fig. 2. The comparison of m_{1k}^{exp} with the theory from Eq. (2) for D and ^{6}Li nuclei; $\alpha_{1k}^{\text{exp}} = 1.46 \pm 0.12$ (run-2001, top) and $\alpha_{1k}^{\text{exp}} = 1.34 \pm 0.08$ (run-2003, bottom) with the opposite polarizations in the cells.

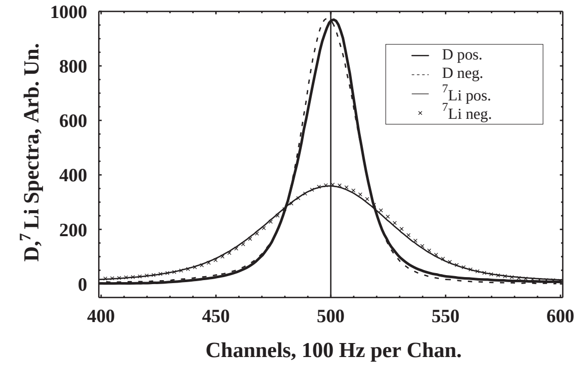

Fig. 3. Comparison of D and ^{7}Li line shapes at the opposite spin temperatures. Spectra of the negative polarization were inverted and aligned with the positive ones; $P_D = \pm 39.2\%$.

Table 2

k	Coils/Year	Field (T)	$\alpha_{1k}^{\text{exp}} \pm \sigma$
D	Inside/2001	2.506	1.44 ± 0.12
^{6}Li	Inside/2001	2.614	1.47 ± 0.10
^{7}Li	Inside/2001	0.9898	1.26 ± 0.11
D	Outside/2003	2.506	1.30 ± 0.08
^{6}Li	Outside/2003	2.614	1.36 ± 0.08
^{7}Li	Outside/2003	0.9898	1.29 ± 0.09
All	Outside/2003	Fit to zero	1.20 ± 0.12

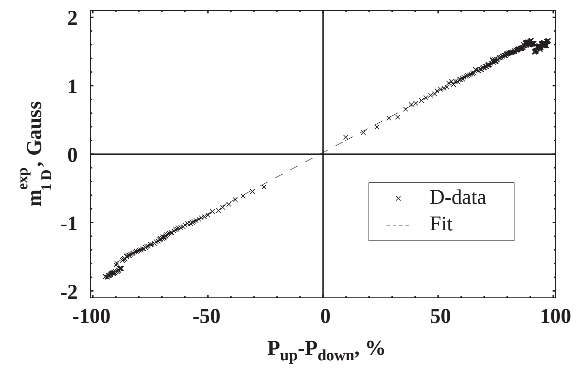

Fig. 4. The shift of the local fields between the oppositely polarized cells as a function of their deuteron polarizations; polarization mode.

the three (of five possible) up, down pairs of central NMR coils in order to reduce the influence of boundary effects on the local field. Table 2 introduces α_{1k}^{exp} obtained from our measurements. Instead of being a constant, α_{1k}^{exp} is decreasing for the nuclei having lower resonant field. This happens because the probing coils have a large size; α_{1k}^{exp} depends on the field gradient which is reduced when the magnetic field decreases.

Another systematic error was found through the comparison of the NMR line shapes of opposite polarizations shown in Fig. 3. One can see that ^{7}Li spectra of opposite polarizations have almost identical line shapes. The analogous D-signals (also ^{6}Li-spectra, not shown here) demonstrate a residual shift between their maxima. This originates from small defects in the cubic lattice leading to the quadrupole interaction of deuterons with the electric field gradient; note that the dipole–dipole interaction leads to a symmetrical broadening around the Larmor frequency. This distortion explains the larger magnitudes of α_{1k}^{exp} in Table 2 for D and ^{6}Li species. The linear fit of all α_{1k}^{exp} from Table 2 to the zero field yields the final result $\alpha_{1k}^{\text{exp}} = 1.20 \pm 0.12$.

The local field in the granulated LiD was measured to be 20% greater than the calculated one for the monocrystalline LiD of the same cylindrical shape.

Fig. 4 shows the field shift between the spectra as a function of the up–down difference of the polarizations when MW are switched on. After switching the MW power off the nuclear spins loose contact with the dipole–dipole reservoir of electron spins and the target goes into the frozen mode. This gives rise to characteristic steps on the curve edges as seen in Fig. 4, which demonstrates the contribution from the electron spins to the first moment of the deuterons [5].

3. The second moment

The experiment shows that the uniform polarization in LiD target can be reached if the nonuniformity of the magnetic field is 0.7 G or lower. Nevertheless, the ultimate negative polarization was about 5% less than the positive one measured in the same probing coil (at the same field inhomogeneity). To understand such a difference, we analysed the second moment of the deuteron line shape using Gaussian approximation. The Gauss line shape is of the form:

$$f(\nu) = f(\nu_0)\exp\left(-\frac{(\nu-\nu_0)^2}{2\Delta^2}\right) \quad (3)$$

where $f(\nu_0)$ represents the amplitude and Δ^2 denotes the second moment in [Hz^2]. The second moment is usually given in field unit [G^2], then with δ denoting the half width at half amplitude of a spectrum, we have:

$$m_{2k} = \frac{\Delta^2}{\gamma_k^2} = \frac{\delta^2}{2\gamma_k^2 \ln 2}. \quad (4)$$

D and ^{7}Li real line shapes and their Gauss fits are shown in Fig. 1.

Fig. 5 shows m_2 of D, ^{6}Li and ^{7}Li as a function of their EST polarizations. The data are fitted by the following equations:

$$m_2(\mathrm{D}) = 1.09 + 2.41(1 - P_{\mathrm{D}}^2), \quad (5)$$

$$m_2(^6\mathrm{Li}) = 0.70 + 2.59(1 - P_{^6\mathrm{Li}}^2) \quad (6)$$

$$m_2(^7\mathrm{Li}) = 2.84 + 0.12(1 - P_{^7\mathrm{Li}}^2). \quad (7)$$

For a monocrystal, the theory predicts that m_2 is given as $m_2 = A - BP^2$ [6] with A and B being constants. The similar form of Eqs. (5)–(7) describe m_2 in the granulated LiD.

In the polarized mode (with the microwave switched on), the evolution of m_2 and of the deuteron polarization over the time are shown in Fig. 6. The comparison of the curves in Fig. 6 indicates that m_2(D) become stepped, when the MW frequency or power are adjusted for the fastest increase of the polarization. It is clear from Figs. 4 and 6 that the contact between electron and nuclear spins through the MW field influences both moments of the NMR line shape.

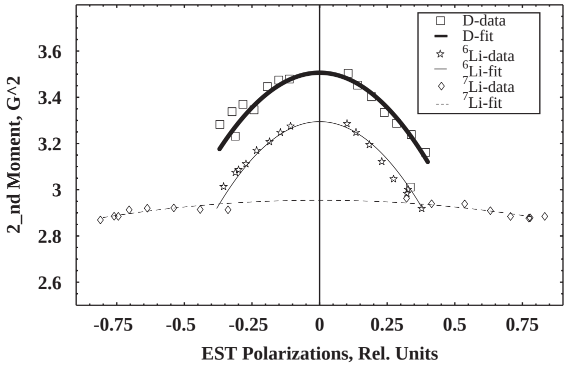

Fig. 5. Comparison of the second moment of D, Li and Li line shapes over their polarization; frozen mode.

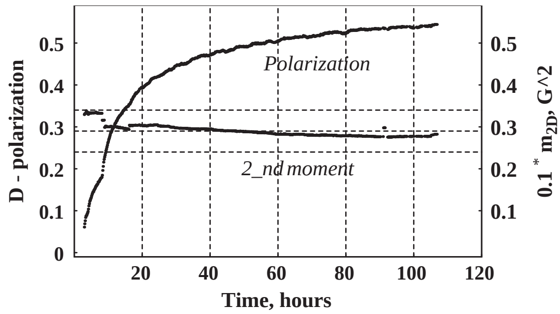

Fig. 6. Evolution of the nuclear polarization and the second moment of deuterons over time; polarization mode.

In turn, the polarized nuclei strongly affect the EPR line. This effect can explain the lower maximum of the negative polarizations in our experiment. In fact, the theory and experiment (see [1, Chapter 6]) show a strong distortion of the EPR line shape in LiH as a consequence of the repopulation within electron spin packages produced by polarized nuclei. Our measurements [7] also detected a considerable shift of 25 G of the EPR line in LiD when the deuterons were

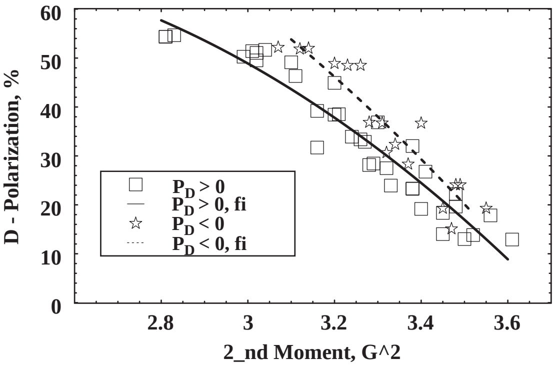

Fig. 7. Module of deuteron polarization as a function of m_2 obtained with the different outer coils distributed along the target; frozen mode.

polarized from zero to 50%. This shift even exceeded the half width of the spectra (about 20 G), therefore the EPR line shape in LiD must be strongly distorted by polarized nuclei.

The difference with LiH is that the EPR line in LiD ($I > \frac{1}{2}$) should be distorted by a different way for opposite nuclear polarizations if there is the quadrupole interactions of nuclei with the electric field gradient. This is indeed the case in LiD. We have demonstrated in Fig. 3 a small contribution of the quadrupole interaction. Well known, the DNP enhancement depends on the width of the EPR line shape [5] which can be detected through the second moment of NMR spectra (see [6, Appendix B]). Following this logic, we have compared, in Fig. 7, the modules of polarizations measured along the target as a function of m_2(D). One can see that the second moment of deuterons tends to be higher for the negative polarization.

The data do not provide a good resolution between the two curves in Fig. 7 because the field is also slightly changing along the target. We assume that the dependence of m_2(D) on the sign of polarization (as shown in Fig. 7) gives an indirect evidence of a slightly different EPR line shapes in LiD of the opposite nuclear polarizations. The fact is that it can qualitatively explain the lower magnitudes of the maximum negative polarizations in our experiments.

4. Conclusion

For the first time the nuclear local field is measured in a granulated LiD material; it was estimated to be 20% larger than given by the calculations for a monocrystalline sample of the cylindrical shape. The second moments of the polarized species in LiD show a qualitative agreement with the theory. We have shown the mutual influence of the electron-nuclei interaction in LiD on the moments of NMR line shapes. The results of our investigation can explain the asymmetry between the maximum positive and the negative polarizations observed in the COMPASS target.

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